

PATENT COOPERATION TREATY

PCT

JC10 Rec'd PCT/PTO 27 JUN 2005

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference RLL-361WO	FOR FURTHER ACTION see Form PCT/ISA/220 as well as, where applicable, item 5 below.	
International application No. PCT/IB2004/002804	International filing date (day/month/year) 30/08/2004	(Earliest) Priority Date (day/month/year) 29/08/2003
Applicant RANBAXY LABORATORIES LIMITED		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 13 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ The international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. ☐ With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, see Box No. I.

2. ☒ **Certain claims were found unsearchable** (See Box II).

3. ☒ **Unity of invention is lacking** (see Box III).

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☐ the text is approved as submitted by the applicant.

☒ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box No. IV. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. With regard to the **drawings**,

a. the figure of the **drawings** to be published with the abstract is Figure No. _____

☐ as suggested by the applicant.

☐ as selected by this Authority, because the applicant failed to suggest a figure.

☐ as selected by this Authority, because this figure better characterizes the invention.

b. ☐ none of the figures is to be published with the abstract.

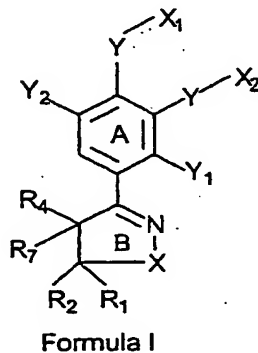
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Box No. IV Text of the abstract (Continuation of item 5 of the first sheet)

The present invention relates to isoxazoline derivatives of structure (I), which can be used as selective inhibitors of phosphodiesterase (PDE) type IV. In particular, compounds disclosed herein can be useful in the treatment of AIDS, asthma, arthritis, bronchitis, chronic obstructive pulmonary disease (COPD), psoriasis, allergic rhinitis, shock, atopic dermatitis, Crohn's disease, adult respiratory distress syndrome (ARDS), eosinophilic granuloma, allergic conjunctivitis, osteoarthritis, ulcerative colitis and other inflammatory diseases in a patient, particularly in humans. The present invention also relates to processes for the preparation of disclosed compounds, as well as pharmaceutical compositions thereof, and their use as phosphodiesterase (PDE) type IV inhibitors.



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A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C07D261/04 C07D413/06 C07D487/08 C07D413/14 C07D498/10
C07D261/20 C07C45/61 C07C269/06 A61K31/422 A61K31/423
A61K31/424

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C07D C07C A61K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, CHEM ABS Data, BIOSIS, EMBASE, BEILSTEIN Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 95/14680 A (PFIZER ; KLEINMAN EDWARD F (US)) 1 June 1995 (1995-06-01) claims 1,15	1-10, 12-31
X	WO 02/50070 A (LEWTHWAITE RUSSELL ANDREW ; KORNBERG BRIAN EDWARD (US); MANNING DAVID) 27 June 2002 (2002-06-27) claim 7 page 134; table 3; compound 18	1-10,13
X	WO 95/14681 A (PFIZER ; KLEINMAN EDWARD F (US)) 1 June 1995 (1995-06-01) page 24 - page 25; compounds 28,29 page 21; compound 21	1-10
	- / - -	



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

8 June 2005

Date of mailing of the international search report

22.06.05

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INTERNATIONAL SEARCH REPORT

 International Application No
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE CAPLUS [Online] CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; FIHI, RACHID ET AL: "Reaction of methylene-.gamma.-butyrolactones with aryl nitrile oxides. Unexpected bisadduct from 5-methylene-5H-furan-2-one" XP002330581 retrieved from STN Database accession no. 1995:376061 compound with RN 162710-40-9 abstract & BULLETIN DES SOCIETES CHIMIQUES BELGES , 104(1), 55-62 CODEN: BSCBAG; ISSN: 0037-9646, 1995, -----	1-8
X	EP 0 515 684 A (CHUGAI SEIYAKU KABUSHIKI KAISHA) 2 December 1992 (1992-12-02) page 13; compounds IV, 28 -----	1-10,13
X	EP 0 387 941 A (JANSSEN PHARMACEUTICA N.V) 19 September 1990 (1990-09-19) claim 1 -----	1-10,13
P,X	TIAN, LI ET AL: "1,3-Dipolar cycloaddition reactions of nitrile oxides to prop-1-ene-1,3-sultone" JOURNAL OF HETEROCYCLIC CHEMISTRY , 40(6), 1071-1074 CODEN: JHTCAD; ISSN: 0022-152X, 2003, XP002330565 table 1; compound 4L -----	1-10,13
X	SALGADO-ZAMORA H ET AL: "OXIDATIVE DEGRADATION OF ARYLFURO-1,2-OXAZOLES TO ARYLNITRILES BY POTASSIUM PERMANGANATE" HETEROCYCLIC COMMUNICATIONS, FREUND PUBLISHING HOUSE, TEL AVIV, IL, vol. 7, no. 3, 2001, pages 209-212, XP009041526 ISSN: 0793-0283 page 210; compound 3A -----	1-10,13
X	HASSIKOU A ET AL: "Synthèse de nouveaux polyhétérocycles" TETRAHEDRON LETTERS, ELSEVIER SCIENCE PUBLISHERS, AMSTERDAM, NL, vol. 42, no. 34, 20 August 2001 (2001-08-20), pages 5857-5861, XP004295942 ISSN: 0040-4039 page 5859; compound 20 ----- -/--	1-10,13

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>SYASSI B ET AL: "Nouvelle méthode de synthèse des 4,5-dihydroisoxazoles en milieu biphasique solide-liquide et par activation ultrasonique" TETRAHEDRON LETTERS, ELSEVIER SCIENCE PUBLISHERS, AMSTERDAM, NL, vol. 40, no. 40, 1 October 1999 (1999-10-01), pages 7205-7209, XP004179018 ISSN: 0040-4039 page 7207; compounds IIE, IIF</p> <p>-----</p>	1-10,13
X	<p>SUBRAMANIAN T ET AL: "FACILE SYNTHESIS OF 3-AROYL-3-SULFOLENES THROUGH CYCLOADDITIONS OF ARYLNITRILE OXIDES & 3-SULFOLENE" SYNTHETIC COMMUNICATIONS, MARCEL DEKKER, INC., BASEL, CH, vol. 27, no. 15, 1997, pages 2557-2562, XP009041534 ISSN: 0039-7911 table 1; compounds 1D, 1F</p> <p>-----</p>	1-10,13
X	<p>BRAHMESHWARI G ET AL: "SYNTHESIS OF 3-ARYLNAPHTHÄ2,3-DÜ-ISOXAZOLE-4,9-DIONES FROM LAWSONE" INDIAN JOURNAL OF CHEMISTRY, SECTION B: ORGANIC, INCL. MEDICINAL, PUBLICATIONS & INFORMATIONS DIRECTORATE, NEW DELHI, IN, vol. 34B, no. 2, February 1995 (1995-02), pages 139-140, XP009041541 ISSN: 0019-5103 compounds 2D,3D</p> <p>-----</p>	1-10,13
X	<p>GABOURY, JANET A. ET AL: "Enantiocontrolled synthesis of burseran, brassilignan, dehydroxycubebin, and other tetrahydrofuran lignans in both enantiomeric forms. Application of intermolecular nitrile oxide cycloadditions and lipase-mediated kinetic resolutions" JOURNAL OF ORGANIC CHEMISTRY, 58(8), 2173-80 CODEN: JOCEAH; ISSN: 0022-3263, 1993, XP002330566 page 2174; compounds 6,9C,9D</p> <p>-----</p> <p>-/--</p>	1-10,13

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>BRAHMESHWARI G ET AL: "SYNTHESIS AND BIOLOGICAL ACIVITY OF FUSED HETEROCYCLES DERIVED FROM EMBELIN"</p> <p>INDIAN JOURNAL OF CHEMISTRY, SECTION B: ORGANIC, INCL. MEDICINAL, PUBLICATIONS & INFORMATIONS DIRECTORATE, NEW DELHI, IN, vol. 30B, no. 3, March 1991 (1991-03), pages 369-370, XP009041539</p> <p>ISSN: 0019-5103</p> <p>table 1; compounds IIID, IVD, VD</p>	1-10,13
X	<p>AWAD W I ET AL: "1,3-DIPOLAR CYCLOADDITION OF NITRILE OXIDES. II. REACTIONS WITH O-QUINOID STRUCTURES"</p> <p>CANADIAN JOURNAL OF CHEMISTRY, NATIONAL RESEARCH COUNCIL. OTTAWA, CA, vol. 47, no. 9, 1969, pages 1473-1477, XP009041540</p> <p>ISSN: 0008-4042</p> <p>page 1475; compound 8C</p>	1-10,13
X	<p>SYASSI B ET AL: "Addition Dipolaire-1,3 des Arylnitriloxydes avec quelques Dipolarophiles Olefiniques sur Alumine en Milieu sec et sous Micro-ondes"</p> <p>TETRAHEDRON LETTERS, ELSEVIER SCIENCE PUBLISHERS, AMSTERDAM, NL, vol. 38, no. 51, 22 December 1997 (1997-12-22), pages 8855-8858, XP004162383</p> <p>ISSN: 0040-4039</p> <p>page 8856; compounds 7G,7H</p>	1-10,13
X	<p>DESCACQ, P. ET AL: "Arylpyrazolines nitrofuraniques: synthèse et propriétés antibactériennes"</p> <p>EUROPEAN JOURNAL OF MEDICINAL CHEMISTRY , 25(3), 285-90 CODEN: EJMCA5; ISSN: 0223-5234, 1990, XP001206364</p> <p>table 1; compounds 3-10</p>	11,13-15
X	<p>BISAGNI, EMILE ET AL: "Synthesis of 3-aryl-4-acetyl-1H-pyrazolo[3,4-b]pyridine s and 3-aryl-4-acetyl-1H-pyrazolo[4,3-c]pyridine s"</p> <p>HETEROCYCLES , 29(9), 1815-24 CODEN: HTCYAM; ISSN: 0385-5414, 1989, XP009048339</p> <p>compounds 4B,5B,16B,17B,</p>	11,13

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>DATABASE CAPLUS [Online] CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; HAIZA, MOHAMMED A.: "Synthesis of some new 3,5-bisaryl-2-pyrazoline derivatives of expected antimicrobial activities" XP002330582 retrieved from STN Database accession no. 1998:725992 compounds with RN 219698-40-5, 219698-37-0, 219698-38-1, 219698-39-2, 219698-41-6, 219698-42-7, 219698-43-8, 219698-47-2 abstract & AL-AZHAR BULLETIN OF SCIENCE , 8(2), 445-454 CODEN: ABSCE7; ISSN: 1110-2535, 1997,</p>	11,13-15
X	<p>----- DATABASE CAPLUS [Online] CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; VARACHE-BERANGER, M. ET AL: "Thienylbenzofurans and arylthienylpyrazolines: synthesis and inhibitory effects on platelet aggregation in vitro" XP002330583 retrieved from STN Database accession no. 1989:608702 compounds with rn 123689-84-9, 123689-85-0, 123689-86-1 abstract & BULLETIN DE LA SOCIETE DE PHARMACIE DE BORDEAUX , 127(1-2-3-4), 37-48 CODEN: BSPBAD; ISSN: 0037-9093, 1988,</p>	11,13-15
X	<p>----- EYNDE VANDEN J J ET AL: "QUATERNARY AMMONIUM SALT-ASSISTED ORGANIC REACTIONS IN WATER: ALKYLATION OF PHENOLS" SYNTHETIC COMMUNICATIONS, MARCEL DEKKER, NEW YORK, NY, US, vol. 31, no. 1, 1 January 2001 (2001-01-01), pages 1-7, XP001019426 ISSN: 0039-7911 figure 1 table 1 ----- -/--</p>	32-34

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	GUAY D ET AL: "Discovery of L-791,943: A potent, selective, non emetic and orally active phosphodiesterase-4 inhibitor" BIOORGANIC AND MEDICINAL CHEMISTRY LETTERS 03 JUN 2002 UNITED KINGDOM, vol. 12, no. 11, 3 June 2002 (2002-06-03), pages 1457-1461, XP002330568 ISSN: 0960-894X	32
Y	page 1458 scheme 1	33,34
X	----- CHAURET N ET AL: "Improving metabolic stability of phosphodiesterase-4 inhibitors containing a substituted catechol: Prevention of reactive intermediate formation and covalent binding" BIOORGANIC AND MEDICINAL CHEMISTRY LETTERS 19 AUG 2002 UNITED KINGDOM, vol. 12, no. 16, 19 August 2002 (2002-08-19), pages 2149-2152, XP002330569 ISSN: 0960-894X	32
Y	page 2150 scheme 2	33,34
X	----- US 5 710 170 A (GUAY ET AL) 20 January 1998 (1998-01-20)	32
Y	column 14 - column 16 column 30; example 9	33,34
Y	----- PATENT ABSTRACTS OF JAPAN vol. 1999, no. 08, 30 June 1999 (1999-06-30) & JP 11 071319 A (ASAHI GLASS CO LTD), 16 March 1999 (1999-03-16) abstract	32-34
Y	----- LANGLOIS B R: "Improvement of the synthesis of aryl difluoromethyl ethers and thioethers by using a solid-liquid phase-transfer technique" JOURNAL OF FLUORINE CHEMISTRY, ELSEVIER SEQUOIA. LAUSANNE, CH, vol. 41, no. 2, November 1988 (1988-11), pages 247-261, XP002232989 ISSN: 0022-1139 page 252; table ii	32-34
Y	----- US 5 712 298 A (AMSCHLER ET AL) 27 January 1998 (1998-01-27) column 10, line 65 - column 11, line 29 ----- -/--	32-34

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P,X	RAJU B ET AL: "Conformationally restricted analogs of deoxynegamycin" BIOORGANIC & MEDICINAL CHEMISTRY LETTERS, vol. 14, no. 12, 21 June 2004 (2004-06-21), pages 3103-3107, XP002330578 ISSN: 0960-894X scheme 6 page 3105	55-62
Y	----- US 4 115 589 A (LEDNICER ET AL) 19 September 1978 (1978-09-19) column 3, line 6 - line 10	55-62
Y	----- VINICK F J ET AL: "AN EFFICIENT SYNTHESIS OF 1 PHENYL-1-PIPERIDINO-TRANS-4-METHYLCYCLOH EXANE UNANTICIPATED TOTAL STEREOSELECTIVITY IN THE CATALYTIC HYDROGENATION OF AN OLEFIN" TETRAHEDRON LETTERS, vol. 28, no. 7, 1987, pages 741-744, XP002330579 ISSN: 0040-4039 page 743; figure	55-62
Y	----- KING FRANK D ET AL: "(Racemic) 3-amino-6-carboxamido-1,2,3,4-tetrahydroca rbazole: A conformationally restricted analogue of 5-carboxamidotryptamine with selectivity for the serotonin 5-HT-1D receptor" JOURNAL OF MEDICINAL CHEMISTRY, vol. 36, no. 13, 1993, pages 1918-1919, XP002330580 ISSN: 0022-2623 scheme 1 page 1918, column 2	55-62
Y	----- NOGUCHI, HIROHIDE ET AL: "Total synthesis and absolute configuration of radiosumin, a strong trypsin inhibitor from the blue-green alga Plectonema radiosum" HETEROCYCLES, 58, 471-504 CODEN: HTCYAM; ISSN: 0385-5414, 2002, XP001206744 scheme 1 page 472 page 473; figure 3 schemes 5,7 page 475 - page 476 -----	55-62

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Box II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☒ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

Although claims 14 and 15 are directed to a method of treatment of the human/animal body, the search has been carried out and based on the alleged effects of the compound/composition.
2. ☐ Claims Nos.:
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

see additional sheet

1. ☒ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☒ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

This International Searching Authority found multiple (groups of) inventions in this international application, as follows:

1. claims: 1-10(part);12-15(part); 16-30; 31(part);

isoxazolines of structure (I) according to claim 1 in which R2 is a linear substituent; corresponding preparation processes and uses

2. claims: 1-10(part); 12-15(part); 31(part); 35-54; 63-74

isoxazolines of structure (I) according to claim 1 in which R1 and R2 together with the isoxazoline ring form a spirocondensed ring system; corresponding preparation processes and uses

3. claims: 1-10(part); 12-15(part)

isoxazolines of structure (I) according to claim 1 in which R2 and R4 together with the isoxazoline ring form a condensed ring system; corresponding preparation processes and uses

4. claims: 11; 12-15(part)

compounds of structure (I) according to claim 11 in which X=S or N, i. e. isozhiazolidines or dihydropyrazoles, and corresponding uses

5. claims: 32-34

a preparation process for benzaldehydes of formulas XXI and XXII

6. claims: 55-62

a preparation process for methylenecycloalkanes according to formula XXXIX

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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Patent document cited in search report		Publication date	Patent family member(s)	Publication date
WO 9514680	A	01-06-1995	AT 187447 T CA 2177375 A1 DE 69422061 D1 DE 69422061 T2 DK 730587 T3 EP 0730587 A1 ES 2139754 T3 FI 945556 A GR 3032506 T3 WO 9514680 A1 JP 2818820 B2 JP 9500146 T PT 730587 T US 5686434 A	15-12-1999 01-06-1995 13-01-2000 30-03-2000 10-04-2000 11-09-1996 16-02-2000 27-05-1995 31-05-2000 01-06-1995 30-10-1998 07-01-1997 31-05-2000 11-11-1997
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Information on patent family members

International Application No

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